SECURING LIQUID FUELS IN THE 21ST CENTURY: ACHIEVEMENTS IN DEVELOPMENT OF THE NEDOL COAL LIQUEFACTION PROCESS

Sadao Wasaka
Director, New Energy and Industrial Technology Development Organization
Tokyo, Japan

ABSTRACT

The New Energy and Industrial Technology Development Organization (NEDO) has been proceeding with development of coal liquefaction technology as part of the "New Sunshine Programs" planned by the Agency of Industrial Science and Technology, a division of Japan's Ministry of International Trade and Industry (MITI). NEDO started basic research for coal liquefaction development in early 1980 and developed the NEDOL coal liquefaction process. The fruit of this development was very successful operation of the 150-t/d pilot plant that was finished in 1998. The results of operation, including engineering data for scaling up the plant to commercial size, were compiled as a technology package. In parallel with the development of coal liquefaction, NEDO is developing product-upgrading technology to make use of these products as transportation fuels. It was found that heteroatoms like nitrogen, sulfur and oxygen can be removed up to the specification levels for gasoline and diesel fuel through hydrogenation. Moreover, the octane number for the upgraded naphtha satisfied the specifications for commercial gasoline, and the upgraded gas oil seems to satisfy commercial specifications for the cetane number for diesel fuel by adding agents improving the cetane number.

1) INTRODUCTION

The research and development program for bituminous coal liquefaction in Japan has been advanced under NEDO's initiative as part of the "New Sunshine Programs" planned by the Agency of Industrial Science and Technology, a division of MITI. The fruit of this development activity was the 150 t/d pilot plant (PP) located in Kashima City, Ibaraki Prefecture, which was operated for two years after 1997 with great results.

NEDO started basic research for coal liquefaction development in early 1980 and developed the NEDOL coal liquefaction process based on the results obtained from this research. NEDO constructed a 1 t/d Process Supporting Unit (PSU) based on the NEDOL concept in Kimitsu City, Chiba Prefecture in 1988, and has been proceeding with research and development. The results from the 1 t/d PSU were incorporated into the design, construction and operation of the 150t/d PP. Through operation of the PP, data for designing large-scale plants was acquired. In parallel with the development of coal liquefaction, development has proceeded on the product upgrading technology that will help make use of these products as transportation fuels. A process development unit (PDU) processing 40 bbl of coal-derived liquid is now under construction, with operation scheduled to start in early 2000.

The results of basic research on the 1-t/d PSU and the 150-t/d PP were compiled as a technology package of the NEDOL process so that they could be utilized to design large-scale coal liquefaction plants. The results from development of product upgrading will be added to the technology package after finishing operation of the PDU.

2) DESCRIPTION OF THE NEDOL PROCESS

Figure 1 shows a process flow diagram of the NEDOL process. The NEDOL process consists of four units: slurry preparation, liquefaction, distillation, and solvent hydrogenation. In slurry preparation, coal is dried and pulverized. Pulverized coal is mixed with recycle solvent and catalyst. This mixture is fed into liquefaction reactors, where coal is liquefied. Products from the reactors are separated into product oils and recycle solvent. The solvent is hydrogenated in a hydrogenation reactor to improve its quality, then recycled to the liquefaction section.

The NEDOL process was developed to produce liquid fuels from coal, and has several features:

1. the process is applicable to a wide range of coals, from sub-bituminous to low-rank bituminous coal;
2. high liquid yield is attained even in mild liquefaction conditions with hydrogen donor solvent and ultra-fine iron catalyst;
3. light distillate yield in liquid products is considerably high; and,
4. stable operation is ensured with reliable process units.

3) DEVELOPMENT RESULTS

The 1 t/d PSU was operated for more than 27,000 hours cumulatively since 1989 and contributed to supporting the 150 t/d PP and improving the NEDOL process. Coals tested in the PP were examined in the PSU, and results from the PSU were used effectively to draw up an operation schedule and determine the operating conditions of the 150 t/d PP.

Construction of the 150 t/d PP was completed in July of 1996, and shakedown operations were conducted for half a year, followed by five operational runs from March 1997 to September 1998. During operation, coals of three different ranks (Tanito-Harum and Adaro coals from Indonesia, and Ikeshima coal from Japan) were liquefied without the need for serious plant modification. In the final run especially, Ikeshima high-rank coal was processed successfully without any intervals after Adaro low-rank coal. This indicates high applicability for coals of the NEDOL process.

The success of the PP operation can be attributed to:

- the reliability of the NEDOL process, which uses a hydrogen donor solvent;
- the reliability of plant design, construction, operation and maintenance; and,
- the support of the PSU, which provided various data to bring about optimum operating conditions for the PP (Reference 1 and 2).

During design and construction of the 150 t/d PP, NEDO determined what targets to achieve, as shown in Table 1, and cleared all of these hurdles during operation. The targets were set to demonstrate that Japanese technology had at least equaled that of Germany and the U.S., which were leading the world in 1980. The targets seemed difficult to obtain considering the limited term and budget of this program (Reference 1), especially attaining more than 54 wt% of liquefied yield, treating 50% of slurry concentration, processing three kinds of coals and operating continuously for more than 1,000 hours.

3.1 Liquefied yields

Table 2 shows the comparison of product yields between the 150 t/d PP and the 1 t/d PSU. The maximum oil yield of the PP was 57.8 wt% (d.a.f. coal base) for Tanito-Harum coal. This yield is the highest value for pilot-scale plants in the world. Note that the yield contains a naphtha fraction (b.p. ~220 C) at more than 60 wt%, thus revealing the characteristics of the NEDOL Process. Meanwhile, the oil yield of the PSU under the same liquefaction conditions was 59.3 wt%. The difference in yield between the PP and the PSU can be explained by the difference in liquid-phase residence time. The PP and PSU have an apparatus that uses the neutron attenuation technique to measure liquid-phase residence time, and the relation between this time and the liquefied yield was measured in each run. Results showed that there is a correlation between slurry-phase residence time and liquefied oil yield as indicated in Figure 2. Longer residence time in the PSU resulted in higher liquid yield than in the PP.

NEDO has developed a "Liquefaction Simulator" for estimating liquefaction yields and hydrodynamics in large-scale reactors. The simulator was used to determine the dimension of large-scale reactors in demonstration plant design by estimating product yields for the reactors.

3.2 Coal concentration

Higher coal concentrations are economically beneficial in that specific coal throughput is improved by processing a greater amounts of coal. Operation with Tanito-Harum and Ikeshima coals achieved a coal concentration of 50 wt%, while Adaro coal was limited to a concentration of 45 wt%. The reason is that the lower ranked coal absorbed more solvent into the fine pores, resulting in higher viscosity of the solvent.

In general, a higher coal concentration results in a lower oil yield. This is caused by a hydrogen shortage per coal given in increased coal throughput to the reactors. During operation of the PSU, it was proved that controlling the quality of the recycle solvent could compensate for the hydrogen shortage and prevent a lower oil yield.

3.3 Kinds of Coals

Table 3 shows coals used in the 1-t/d PSU and the 150-t/d PP. In the PSU, nine kinds of coal ranging from sub-bituminous to low-rank bituminous were examined. Based on the experiments, three kinds of coal were selected for use in operation of the PP. Processing a wide range of coals clarified the effects of the coals on slurry rheology, reaction heat, and reaction kinetics.

In the final run especially, Ikeshima high-rank coal was processed successfully without any intervals after Adaro low-rank coal. This indicates high applicability for coals of the NEDOL process.

3.4 Product characteristics and upgrading

Coal-derived liquid generally includes a relatively large amount of heteroatoms such as nitrogen, sulfur and oxygen, and requires further processing for use as transportation fuel. Table 4 shows the characteristics of crude liquefied oil from the NEDOL process. NEDO has been conducting research and development into product upgrading to make use of these products as transportation fuels in parallel with development of the liquefaction process. The target of development is to produce gasoline and diesel fuel that satisfies commercial specifications while improving the octane number for gasoline, and the cetane number for diesel fuel.

Use of the naphtha fraction as transportation fuels requires a catalytic reforming process to improve the octane number. Because olefins and heteroatoms in the fraction react as catalytic poisons during the process, these compounds have to be eliminated beforehand. It was proved that these compounds are eliminated through hydrotreatment, and the octane number of gasoline improves via the catalytic reforming process as shown in Table 5.

Improvement of the cetane number is required for use of the gas-oil fraction as diesel fuel. The gas-oil fraction of the coal-derived liquid includes higher amounts of aromatic compounds, and the cetane number is lower than that of petroleum gas oil. The cetane number of petroleum gas oil can easily be improved by hydrotreatment. However, it was difficult to attain a cetane number greater than 40 for the coal-derived gas oil, even in the more severe conditions of hydrotreatment; a cetane number of only 38 was attained in the optimized conditions shown in Table 6. Addition of the agent to improve the cetane number seems to be a good practical procedure.

NEDO was constructing a PDU to process 40 bbl of coal-derived liquid a day. Construction was scheduled for completion by the end of 1999, and operation of the plant by April 2000. Engineering data for scaling up the plant will be acquired through operation. Figure 3 shows the outline of the plant. Coal-derived liquid from the liquefaction plant is hydrotreated and separated into naphtha, kerosene and gas oil. Naphtha is hydrotreated and reformed for gasoline. Gas oil is hydrotreated for diesel fuel.

4) TECHNOLOGY PACKAGE

NEDO has compiled all the data obtained from the PP and the PSU as a technology package that addresses the reliability, economics, and environmental integrity of the technology. The structure of the package is shown in Figure 4. It is composed of: 1) Engineering Data; 2) Scale-up Data; 3) Operation & Maintenance Data; 4) Simulation; 5) Conceptual Design; and 6) Economic Study.

The data acquisition plan for each operation of the PP was designed in advance. Operations were carried out according to the plan, and data for the package were acquired efficiently. These data made it possible to correlate the data between the PP and the PSU, for example, on newly developed equipment such as the liquefaction reactor. Correlation of data provided a tool that can be used for scale-up of the 1 PP to a demonstration plant. Data on gas hold-up in the reactor, heat of reaction, and the heat recovery system were compiled to a great extent numerically.

The items were determined mostly at the design stage of the PP, and some were incorporated throughout operation. Operation of the PP, in turn, provided a great deal of meaningful knowledge that reflected on the conceptual design of a demonstration plant.

5) PLANS FOR COMMERCIALIZATION

China and Indonesia are supposed to experience a shortage of transportation fuels in the near future (Reference 3). Coal liquefaction seems to be a good solution for providing liquid fuels, especially to China and Indonesia, which have enormous coal resources. Since the oil crises, the United States and Germany have been developing coal

Proceedings of the Advanced Clean Coal Technology International Symposium 1998. Tokyo, Japan; pp. 198-207.

Yoshida, H. 1998. "Development Results and Future Aspects of NEDOL Proce *Final Report of Coal Liquefaction Pilot Plant Operation.* Nippon Coal Oil Co., Ltd.; pp 1-11.

Jialu, Jin. 1998. "Status of Energy Sources and Commercialized Prospect of Direct Coal Liquefaction Technology in China." *Proceedings of the Advanced Clean Coal Technology International Symposium 1998*. Tokyo, Japan; pp. 255-264.

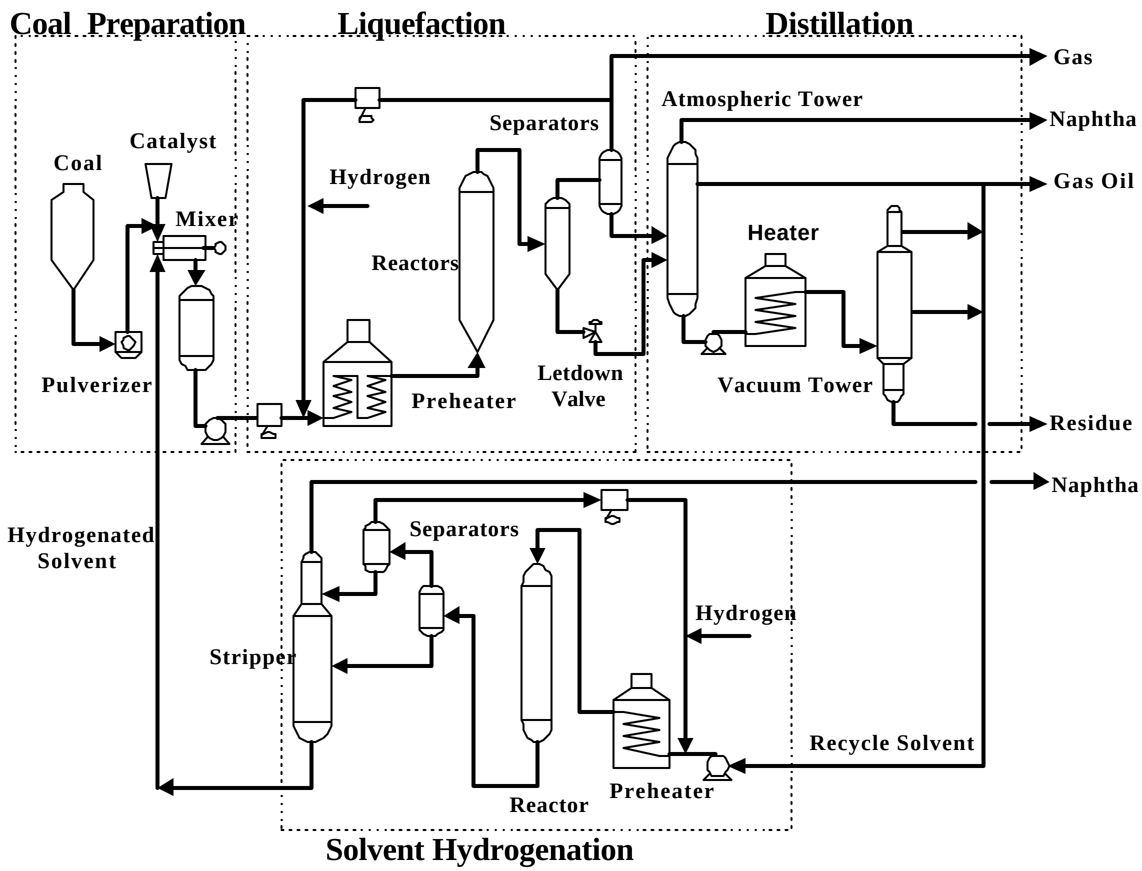

Figure 1 NEDOL Process Flow Diagram

Table 1 Targets and Results of the Pilot Plant

Item	Targets	Results	Evaluation
1. Oil Yield	> 54wt% (630 l/t-coal or 4 bbl/t-coal)	58 wt%	
2. Slurry Concentration	40 -50wt%	50 wt%	
3. Coal Rank	From sub-bituminous coal to low ranked bituminous coal (3 kinds)	3 kinds	
4. Continuous Operation	> 1,000 hrs	1,920 hrs	
5. Amount of Liquefaction Catalyst	2 - 3 wt% / coal	1.5 wt%	
6. Improvement of Thermal Efficiency	Heat recovery with slurry heat exchangers	Save 60% of heat in preheater	
7. Performance of Newly Developed Equipment and Materials	Demonstrate in the PP	Demonstrated in the PP completely	
8. Technology Package	Compile all data into Technology Package	Compiled successfully	

Table 2 Comparison of Product Yield between PP and PSU
(Tanito Harum Coal , Natural Pyrite Catalyst)

		Standard Condition		Maximum Condition	
		PP	PSU	PP	PSU
Gas Yield	(wt% , daf)	17.6	19.7	21.4	26.5
Oil Yield	(wt% , daf)	50.7	51.7	57.9	59.3
Residue	(wt% , daf)	26.1	23.3	16.0	9.9
Actual Residence Time	(min)	86	99	-	161

PP : Pilot Plant , PSU : Process Supporting Unit

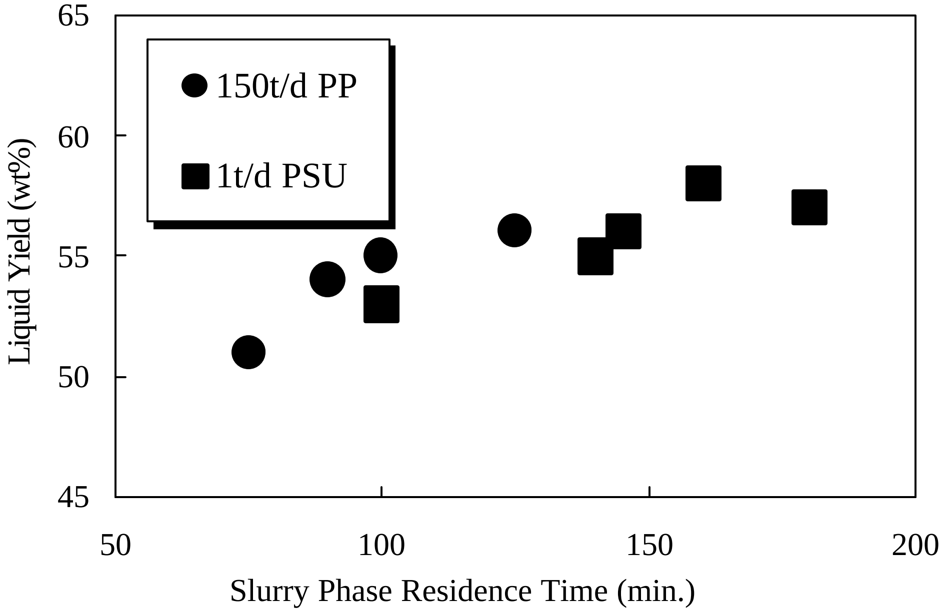

Figure 2 Relationship Between ResidenceTime and Liquid Yield

Table 3 Coals in the NEDOL process

Carbon content (wt%, daf coal)	72	73	74	75	76	77	78	79	80	81	82	83	84
Coal		o Wyoming (USA)	• Adaro (Indonesia)		• Tanito Harum (Indonesia)	o Taiheiyo (Japan); o Wandoan (Australia)		o Yilan (China)	o Illinois#6 (USA)	o Shenhua (China)	• Ikeshima (Japan)		
Rank		Low rank sub-bit.				Sub-bit.		Low rank bit.				Bit.	

Coal for NEDOL Process

o: tested at 1t/d PSU

•: tested at 1 t/d PSU and 150 t/d PP

Table 4 Characteristics of Coal Derived Liquid (Tanito Harum coal from Indonesia)				
			Crude	Primary Hydrogenated
Specific Gravity (g/cm^3)			0.8994	0.8204
Elemental Analysis				
	C	(wt%)	84.7	
	H	(wt%)	10.9	
	N	(ppm)	6400	1
	S	(ppm)	500	5
	O	(wt%)	3.5	
Composition				
	-180	(vol%)	18.3	35.8
	180 - 240	(vol%)	40.2	39.2
	240 - 340	(vol%)	41.5	25.0

Table 5 Characteristics of Naphtha (Wandoan coal from Australia)					
			Crude	hydrotreated	Reformate
Specific Gravity (g/cm^3)			0.791	0.763	0.818
Octane Number		RON		53	97.4
Composition					
	Saturated	(vol%)	78.0	90.4	35.8
	Olefins		10.4	0.1	0.5
	Aromatics		11.6	9.5	63.7
Elemental Analysis					
	C	(wt%)	83.7		88.2
	H	(wt%)	13.1		11.1
	N	(ppm)	1800	<1	
	S	(ppm)	1000	1	
	O	(wt%)	3.4		

Table 6 Characteristics of Diesel Fuel (Wandoan coal from Australia)				
			Crude	Hydrotreated
Specific Gravity		(g/cm^3)	0.9398	0.8819
Cetane Number			24.8	38
Elemental Analysis				
	C	(wt%)	87.9	86.3
	H	(wt%)	10.9	13.2
	N	(ppm)	1380	11
	S	(ppm)	56	<1
	O	(wt%)	1.2	<0.5

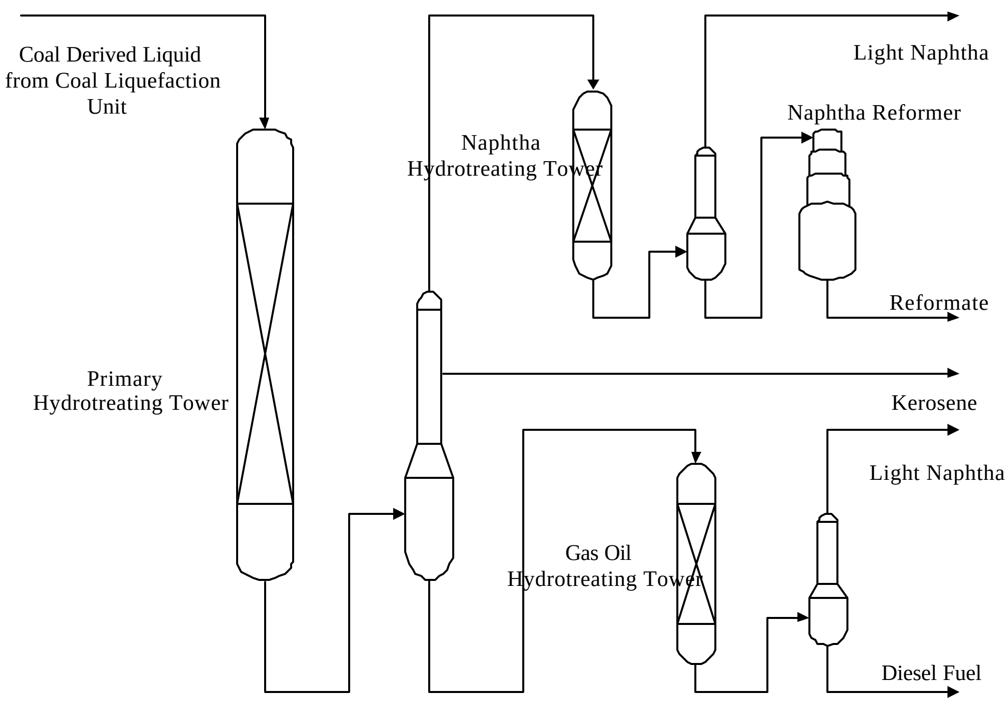

Figure 3 Conceptual Flow for Coal Derived Liquid Upgrading

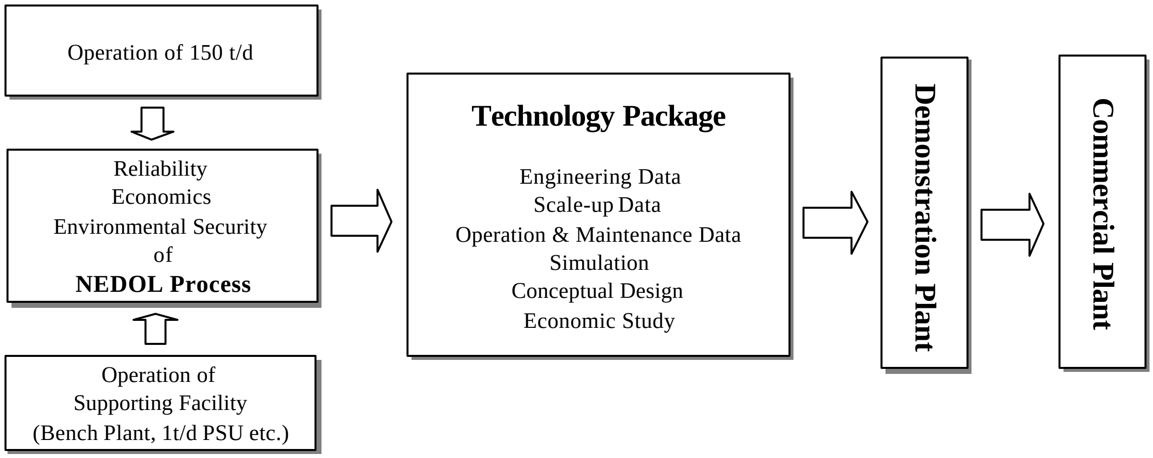

Figure 4 NEDOL Process Technology Package